Silver Content of Precipitation from Seeded and Nonseeded Florida Cumuli

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ABSTRACT

A subprogram of NOAA's 1973 Florida Area Cumulus Experiment (FACE) was undertaken to determine the silver content of precipitation associated with convective clouds massively seeded with silver iodide nucleant over southern Florida. An atomic absorption analysis of 127 rainwater samples collected just below cloud base by a polypropylene-lined scoop mounted on the fuselage of the NOAA DC-6 aircraft indicated that the mean concentration of silver obtained on seed days (69 samples) was no greater (and, in fact, appreciably less) than that obtained on no-seed days (58 samples). In both sets of samples, the median concentration of silver was more than two orders of magnitude lower than the U. S. Public Health safety limit of 5×10^{-8} g ml^{-1}. Of the 69 aircraft samples collected on seed days, only two contained a concentration of silver in excess of 1×10^{-9} g ml^{-1}. Of the 58 aircraft samples collected on no-seed days, eight contained silver in concentrations exceeding 1×10^{-9} g ml^{-1}. The samples collected from the aircraft showed higher mean concentrations of silver than did those collected on the ground. An atomic absorption analysis of 79 rainwater samples collected at both fixed and mobile sites on the surface showed that the mean concentration of silver on seed days was three orders of magnitude less than 5×10^{-8} g ml^{-1}; the maximum concentration of silver found in any sample did not exceed 1×10^{-9} g ml^{-1}. Statistical results from the nonparametric Mann-Whitney Wilcoxon test suggest that the surface seeded (34 samples) and surface nonseeded (45 samples) data come from the same population (i.e., no significant differences between the two data sets). There is some evidence (from a separate set of surface rainwater samples collected upwind of the target area) to suggest a persistently higher (by about a factor of 2 or 3) mean concentration of silver during the course of the experiment than either before or after the experiment.

1. Introduction

The main objective of NOAA's 1973 Florida Area Cumulus Experiment (FACE) was to seed groups of supercooled tropical cumulus clouds to convert enough water to ice so that sufficient heat would be released to enhance cloud growth and augment rainfall over a specific area. It is hypothesized that massive seeding with silver iodide can, under prespecified conditions, alter cloud dynamics to produce well-organized and long-lasting convective systems capable of efficiently processing the available low-level moisture. The design and execution of the experiment has been described in detail (Woodley and Sax, 1976).

FACE 1973 was conducted during 94 days from mid-June through late September. Concurrent with a core program designed to measure the effects of "dynamic" seeding on area-wide rainfall, a subprogram to determine the silver concentration of precipitation from seeded and nonseeded cumuli was carried out. Bulk rainwater samples were collected from the NOAA DC-6 aircraft just below cloud base, and from fixed and mobile sites at the surface. The 13 000 km^2 experimental area is shown in Fig. 1. Although the aircraft was free to collect rainwater anywhere within the large quadrilateral, the surface collection sites were located mainly within the stippled "intensive network" area.

The seeding decision was randomized by day with *all* selected clouds on a given "GO" day either seeded or not seeded. On a seeding GO day, 60–400 pyrotechnics, each emitting 50 g of silver iodide, were released at the -10°C isotherm level within the active updraft regions of suitable cumulus towers. Because of the large amount of silver iodide often released on a given GO day, it is important to consider the ecological and environmental impacts of the experiment. The drinking water standards from the U. S. Public Health Service list a mandatory silver limit of 5×10^{-8} g ml^{-1}. Studies have shown

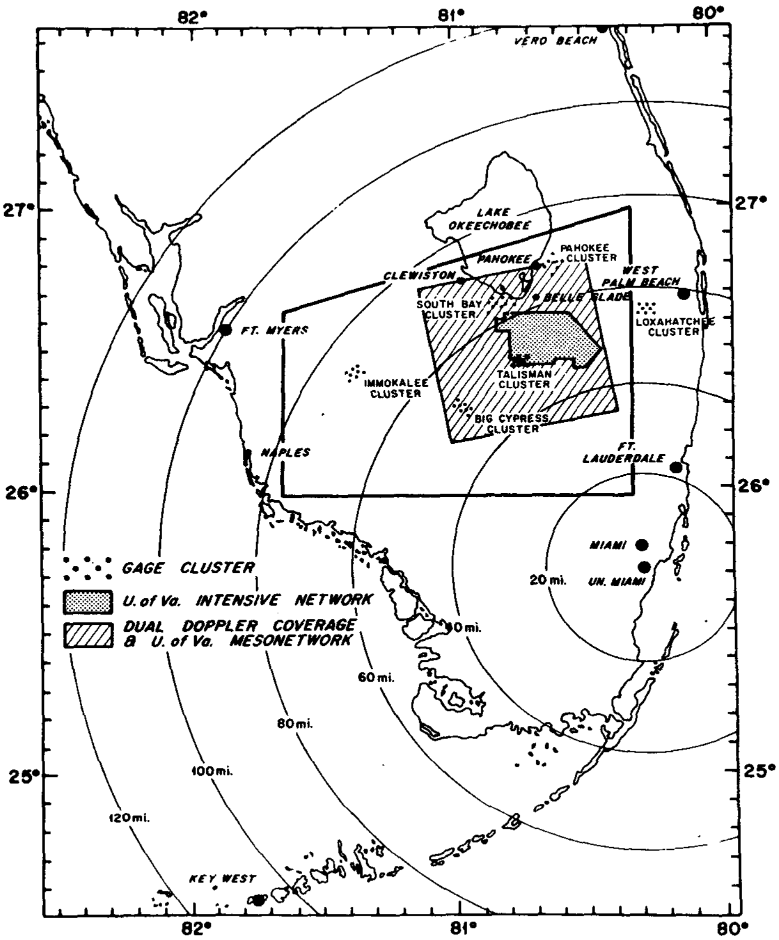

Fig. 1. FACE 1973 target area. Stippled area represents region of highly concentrated meteorological instrumentation and is where the surface rainwater collection program was primarily conducted.

that silver above this concentration can produce argyria in humans, a condition manifested by a bluish, permanent darkening of the skin. McKee and Wolf (1963) have shown that concentrations above 4×10^{-9} g ml^{-1} are toxic for certain fish and other lower organisms. A comprehensive review of the ecological effects of silver iodide has been documented by Cooper and Jolly (1970).

2. Procedure

Analysis of silver content in precipitation collected on the ground during seeding experiments has been performed by several groups (Warburton and Young, 1968; Warburton, 1973; Parungo and Robertson, 1969; Summers, 1972). In addition, Ostlund and Stearns (1970) used an aircraft to collect rainwater for subsequent analysis of silver content under and within seeded complexes. The design of the FACE 1973 silver sampling program was inspired to a large extent by the results, conclusions and the problems of the above programs.

Approximately 280 rainwater samples were collected during the FACE 1973 program.[1] Half of these samples was collected at the surface in 400 ml polypropylene beakers. The other half of the samples was collected aboard the NOAA DC-6 aircraft just below the cloud base level (600 m) with a scoop mounted on the top front portion of the aircraft fuselage (Fig. 2). The scoop was lined with polypropylene, a material that was tested and shown to present no significant silver adsorption problems. The DC-6 aircraft, vectored into position underneath the cloud system being seeded[2] by the upper-level (6 km) C-130 aircraft, carried out repeated penetrations on reciprocal headings through the rainshaft. With the scoop presenting a cross-sectional area of approximately 100 cm^2, it was possible to sample about 20 m^3 of rain volume and to collect about

[1] Some low-volume samples collected during several traverses through very light precipitation were combined into one; this resulted in a total of 258 samples available for analysis.

[2] Seeding action was either real or simulated depending upon randomized decision for clouds on that day.

TABLE 1. Silver concentrations ($\times 10^{12}$ g ml^{-1}) for all aircraft and surface samples collected within the 13 000 km² seeding network.

Group	Number of samples	Mean	Standard deviation	Standard error	Median	Maximum	Minimum
Total aircraft	127	4880	31100	2750	92	300000	BD**
Total aircraft*	123	320	964	87	92	7200	BD
Seeded aircraft	69	2190	16800	2030	46	140000	14
Seeded aircraft*	68	160	386	47	46	3000	14
Nonseeded aircraft	58	8070	42100	5530	130	300000	BD
Nonseeded aircraft*	55	518	1357	183	130	7200	BD
Total surface	79	58	131	15	24	970	BD
Seeded surface	34	68	170	29	37	970	BD
Nonseeded surface	45	52	95	14	20	540	BD
Mobile seeded	24	43	65	13	36	320	BD
Mobile nonseeded	32	45	64	11	22	320	BD
Fixed seeded	10	130	30	94	13	970	BD
Fixed nonseeded	13	66	148	41	23	540	BD

* Data excluding one seed and three no-seed samples showing anomalously high concentrations of silver (see text for discussion).
** BD signifies a level below detection limit of the instrument.

30 ml of rainwater on each traverse through moderate or heavy precipitation. The aircraft samples were frozen immediately in dry ice. Air-to-ground communications made possible the mobile collection of rainwater at the surface, and about 70% of the network surface samples were collected in this manner. On these occasions a van was maneuvered underneath the clouds being seeded, and a 400 ml beaker was mounted on a tripod and exposed. The remaining 30% of the surface samples were collected at fixed sites within the stippled region shown in Fig. 1. The surface samples were frozen as soon as possible, usually within 4–6 h after collection. All samples were kept frozen until analysis.

After being allowed to melt, the rainwater samples were analyzed[3] with a Perkin Elmer 403 flameless atomic absorption spectrometer equipped with a HGA-70 heated graphite atomizer and a deuterium arc background corrector. The instrumentation and analysis technique have been fully described by Segar and Gilio (1973). The absolute detection limit for silver using the technique is approximately 8×10^{-13} g ml^{-1}, which represents a considerable improvement in detection threshold over analysis techniques cited previously in the literature.

[3] Analysis was supervised by Dr. Doug Segar at the University of Miami's Rosenstiel School of Marine and Atmospheric Science.

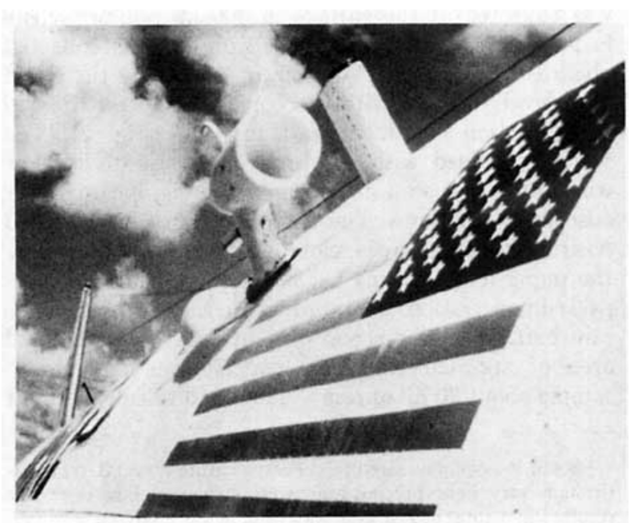

FIG. 2. Polypropylene scoop used for collecting rainwater shown mounted in position on NOAA DC-6 fuselage.

3. Results

A summary of the samples collected and analytical results are given in Tables 1–4. Each of the tables contains a description of the sample set of interest, the number of samples collected within each set, the mean silver concentration, and the standard deviation, standard error of the mean, and the median, maximum and minimum silver concentrations within each set. All silver concentrations are expressed in units of 10^{12} g ml^{-1}.

Table 1 gives the results for all aircraft and surface samples collected within the 13 000 km² seeding network. The table is subdivided into two main groups, aircraft-collected samples and surface-collected samples, the latter of which is further subdivided into samples collected from the mobile van and samples collected at the fixed sites.

Table 2 gives a stratification of the aircraft samples by date of collection. The first grouping lists the four dates the aircraft collected samples on seed days and the second lists six dates the aircraft collected samples on no-seed days. The final category contains ten samples that were collected in a Hurricane Delia research flight, well removed in space and time from any seeding activity.

Table 3 gives the breakdown of samples collected on the roof of the NOAA National Hurricane Center in Coral Gables, a site ~25 mi south of the southeastern edge of the network and usually upwind (at low- and

Table 2. Silver concentration ($\times 10^{12}$ g ml^{-1}) for specific aircraft collection dates.

Group	Number of samples	Mean	Standard deviation	Standard error	Median	Maximum	Minimum
Aircraft seed dates							
25 July	23	107	127	27	37	430	14
12 August	7	20300	52800	20000	320	140000	71
12 August*	6	306	264	108	260	790	71
13 August	13	100	70	19	71	230	32
12 September	26	202	598	117	36	3000	14
Aircraft nonseed dates							
2 August	7	43000	113000	42800	94	300000	21
2 August*	6	113	131	54	77	370	21
4 August	4	104	41	20	97	150	55
6 August	8	4170	7160	2530	210	20000	74
6 August*	7	622	1103	417	160	3100	74
8 August	7	233	218	83	140	710	81
9 August	16	8600	29800	7440	340	120000	83
9 August*	15	1370	2314	598	280	7200	83
9 September	16	53	62	16	21	220	BD
Aircraft hurricane dates							
3 September	10	45	48	15	26	100	BD

* Data excluding one seed and three no-seed samples showing anomalously high concentrations of silver (see text for discussion).

mid-tropospheric levels) of seeding operations. The table is grouped into total collected samples and samples collected before, during and 1 week following seeding activities.

Table 4 stratifies the results of the silver analysis on the basis of collected volume for all aircraft samples (with the exception of 25 for which volumes were not recorded). The samples, which were collected in 120 ml bottles, are grouped in the table as those collected with volumes greater than 30 ml and those collected with volumes 30 ml or less. The samples are subdivided into three categories: total number of samples, seeded samples and nonseeded samples.

4. Discussion

The mean surface silver concentrations shown in Table 1 compare favorably with other surface collection programs cited earlier in the paper. The mean seeded surface silver concentration of 6.8×10^{-11} g ml^{-1} is slightly higher than the mean nonseeded surface silver concentration of 5.2×10^{-11} g ml^{-1}. These differences are not at all significant based upon results from the nonparametric Mann-Whitney Wilcoxon test,[4] a finding which suggests that all data were derived from a common population.

The most perplexing series of results concerns the aircraft silver concentrations. The aircraft-obtained data showed that the nonseeded silver mean of 8.1×10^{-9} g ml^{-1} greatly exceeded the seeded silver mean of 2.2×10^{-9} g ml^{-1}, these differences being significant at the 99.5% significant level using the Mann-Whitney Wilcoxon test. With the complete data set considered, the aircraft silver means exceed the surface silver means by almost two orders of magnitude. Any attempts to directly compare the aircraft and surface results, however, must be made with great caution because the sample sets were obtained through different collection methods and, perhaps more importantly, because there

[4] The Mann-Whitney Wilcoxon test is a nonparametric statistical method for testing the null hypothesis that two independent samples come from identical continuous populations; for details the reader is referred to engineering statistics texts such as *Introductory Engineering Statistics* by Irwin Guttman and S. S. Wilks, Wiley and Sons, 1965, p. 231.

Table 3. Silver concentrations ($\times 10^{12}$ g ml^{-1}) for Coral Gables.

Group	Number of samples	Mean	Standard deviation	Standard error	Median	Maximum	Minimum
Before experiment	2	23	4	3	23	26	BD
After experiment	9	14	9	3	12	28	20
Before and after	11	16	9	3	14	28	BD
During experiment	31	55	74	13	42	350	BD
Total, all samples	42	46	66	10	26	350	BD

TABLE 4. Aircraft silver concentration ($\times 10^{12}$ g ml^{-1}) stratified by collection volume.

Group	Number of samples	Mean	Standard deviation	Standard error	Median	Maximum	Minimum
All > 30 ml	44	264	1000	150	67	6600	BD
All ⩽ 30 ml	58	10432	45531	5979	170	300000	BD
All ⩽ 30 ml*	54	464	1129	154	170	7200	BD
Seeded > 30 ml	16	44	37	9	32	160	14
Seeded ⩽ 30 ml	28	5268	26411	499	115	140000	BD
Seeded ⩽ 30 ml*	27	649	1477	284	115	3000	BD
Non-seeded > 30 ml	28	391	1244	235	115	6600	BD
Non-seeded ⩽ 30 ml	30	15250	58099	10607	205	300000	BD
Non-seeded ⩽ 30 ml*	27	278	587	113	205	7200	BD

* Data exluding one seed and three no-seed samples showing anomalously high concentrations of silver (see text for discussion).

were no representative dates of surface sampling that coincided with any of the 10 aircraft dates.

It can also be observed from Table 1 that the median values of silver concentration differ much less between aircraft- and surface-collected samples than do the mean values, particularly in the case of the seeded sample sets. This suggests that the very high mean concentrations of silver collected by the aircraft may be due to several "anomalously or extraneously" high data points. Indeed, an examination of the raw data showed that only two of the 69 aircraft seeded samples contained silver in concentrations exceeding 1×10^{-9} g ml^{-1}, with one of these values exceeding 1×10^{-7} g ml^{-1}. In the aircraft nonseeded sample set, eight values of silver concentration in excess of 1×10^{-9} g ml^{-1} were catalogued, with three of those exceeding 1×10^{-8} g ml^{-1}. The data in Tables 1, 2 and 4 have been additionally stratified to exclude all four silver concentration values in excess of 1×10^{-8} g ml^{-1} and although these results still show the mean concentrations of silver collected by the aircraft to be higher than those collected on the ground, the differences are in the range of about a factor of 3–10 instead of the more than two orders of magnitude found by considering the total data set. It is thus possible that the very few exceptionally high ($>1\times10^{-8}$ g ml^{-1}) concentrations of silver were caused by contamination problems in preparing the samples for analysis, although this cannot be proven definitely and other explanations are also plausible. The very high silver concentrations excluded in the stratified grouping comprise less than 2% of the total data set analyzed.

The overall higher aircraft silver concentrations do not appear to be the result of contamination from residual silver in the scoop. The aircraft scoop and tubing were washed with nitric acid and rinsed several times with distilled water before each flight. The last category in Table 2 shows the average silver concentration of ten samples collected in a Hurricane Delia research flight. These samples were collected over the Gulf of Mexico on 3 September 1973, far removed in space and time from any Florida seeding activity. The DC-6 for much of the two weeks before this flight had been used as the primary seeder aircraft for the FACE program. If residual silver contamination were an important factor, it would have shown up in these samples. The mean of these samples is 4.4×10^{-11} g ml^{-1}, considerably less than the south Florida aircraft silver concentration mean. On the strength of these results, it would appear that the relatively high aircraft silver concentrations over the FACE target area reflect actual conditions at the time and place of sampling.

The overall higher aircraft silver concentrations could be a result of the aircraft scoop design. The scoop has a major deficiency common to most rain collectors. It collects aerosol and rainwater together, and it is impossible to separate the particulate matter of the two components. It is difficult to estimate the relative concentrations of aerosol being collected because the scoop is always open to the airstream, both within and outside the showers. Wisniewski *et al.* (1974) collected aerosol samples simultaneously with the rainwater samples aboard the aircraft. Analysis of these samples by x-ray spectroscopy techniques showed silver particulate matter to be present in the atmosphere, which may help to explain the overall higher aircraft silver concentrations. Some support for this suggestion can be found in a study by Rancitelli and Perkins (1970) who sampled aerosol in multiple layers over the western United States. They found the silver concentrations to vary by four orders of magnitude between altitudes of 3 and 15 km. The highest concentration was found at 8.8 km. They hypothesized these high concentrations to be the result of industrial air pollution and/or cloud seeding activities.

The contamination of no-seed data due to industrial air pollution at first appears to be unlikely since the air mass over south Florida at mid- to low-tropospheric levels in summertime is usually of maritime tropical origin. As illustrated in Figs. 3 and 4, however, the air mass on many days at the 200 mb (~12.4 km) level originated in the continental United States. Note particularly the trajectory analyses for 25 July, 8 August, and 9 and 12 September. Thus, it is entirely possible

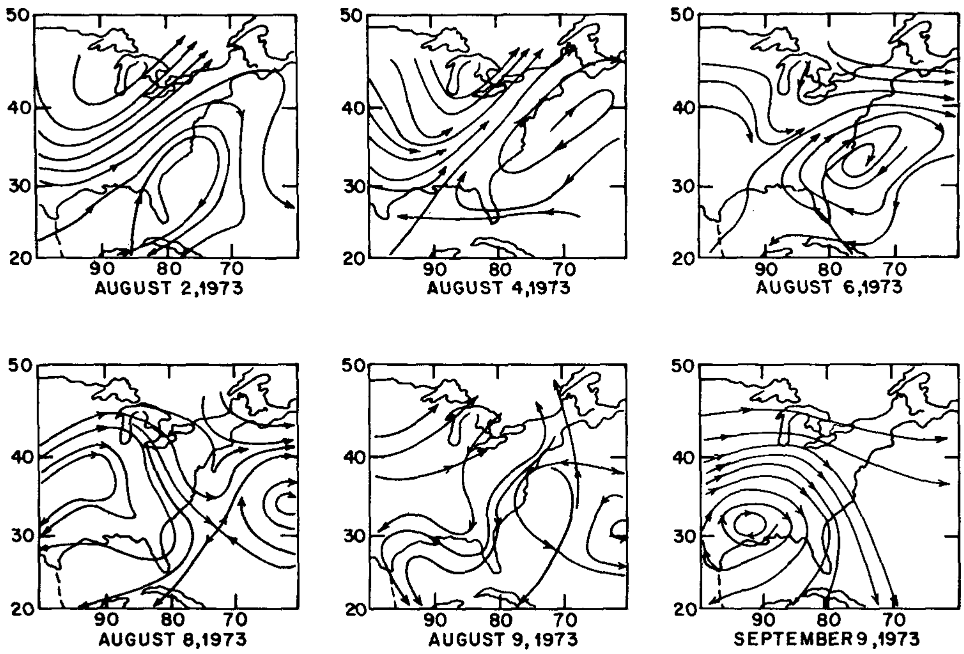

FIG. 3. 200 mb trajectory analyses at 1200 GMT for six nonseeded days during FACE 1973.

that some high concentrations of silver in rainwater could have been a result of the entrainment and wash-out of industrial air pollution existing at upper levels. Since such a process would require that convection penetrate these levels, it would be expected to be dependent upon both the intensity and duration of the convective systems. A far more exhaustive background study is needed to resolve this question, however.

The contamination of nonseeded data because of persistence of silver iodide from previous seed days

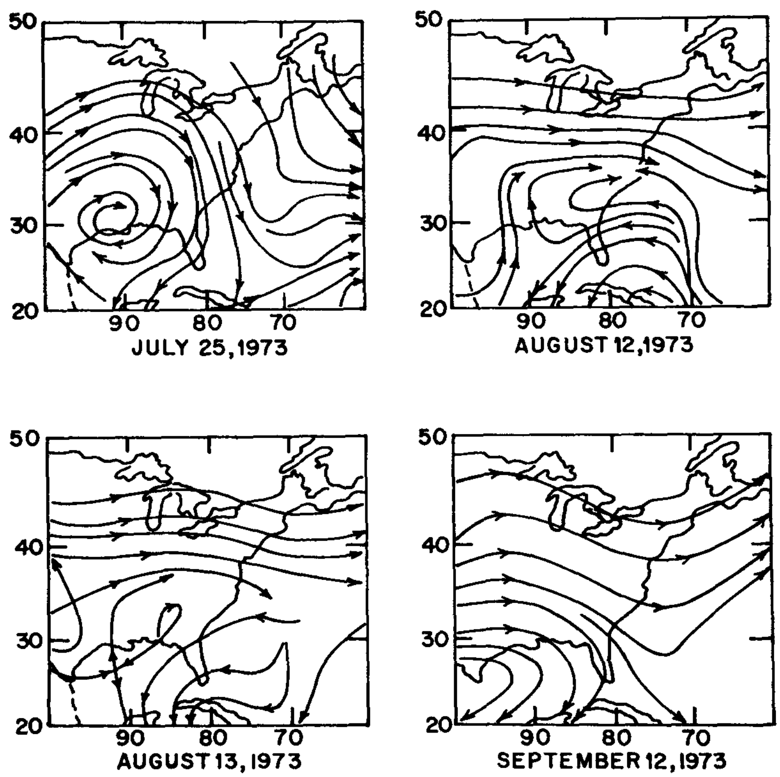

FIG. 4. 200 mb trajectory analyses at 1200 GMT for four seeded days during FACE 73.

may also be a factor. Samples were collected from early July through early October 1973 on the roof of the NOAA National Hurricane Center in Coral Gables, which (as noted earlier) is usually upwind of seeding operations. The mean of the 42 samples analyzed was only 4.5×10^{-11} g ml^{-1}. When the data are further stratified, however, some interesting results appear. The data are subdivided into three categories as shown in Table 3: samples collected before the first seeding of 7 July, samples collected from 7 July until 19 September (one week after the last seeding which took place on 12 September), and samples collected after 19 September. The one-week period following the conclusion of seeding activity was picked arbitrarily, considering the site's location upwind of the network, as a time after which any persistence effects from the seeding could be ignored. When the silver concentrations of the 31 samples collected during the 7 July–19 September period are compared to the 11 samples collected before and after this period, the differences in mean silver concentration are significant at the 99.5 significance level (using the Mann-Whitney Wilcoxon test). This suggests that the higher silver concentrations found during the project at the Coral Gables site resulted from contamination and persistence from previous seeding activities, but further background data are needed.

Gatz (1975) has noted that the highest background concentrations of silver in Illinois rainwater are generally associated with light rainfalls. Results from the stratification of aircraft data by collection volume shown in Table 4 indicate that small-volume seeded samples ($\leqslant$30 ml) contain considerably higher mean concentrations of silver than do large-volume seeded samples. These differences within the aircraft-seeded data set were found to be significant at better than the 99.5 level using the Mann-Whitney Wilcoxon test. With the three "extraneously" high nonseeded data points removed, however, no significant differences are observed when the nonseeded data set is stratified by volume. Further attempts to stratify the data on the basis of specific collection dates (seeded versus nonseeded) and cloud types (small relatively isolated convective clouds versus moderately large cloud complexes) yielded no clear-cut trends in the data.

It should be mentioned here that the spatial and temporal problems associated with penetrating rainshafts of convective clouds seeded 5 km above the sampling altitude are considerable and form the subject of a companion paper (Wisniewski and Cotton, 1976). Because of other data collection requirements levied on the DC-6 during FACE 1973, only rarely was the aircraft maneuvered directly beneath a subject cloud during the exact interval of penetrations and treatment by the C-130. Therefore, with the exception of a few case studies discussed by Wisniewski and Cotton (1976), it can only be claimed that water was collected on seed and no-seed days, and it is not known with certainty that all rainshafts sampled can be expected to have contained the products of a seeding treatment at the times they were penetrated.

5. Conclusions

A subprogram of NOAA's 1973 Florida Area Cumulus Experiment (FACE) was conducted to determine how seeding with silver iodide affected the silver content of south Florida convective precipitation. Rainwater was collected just below cloud base level (127 samples) and on the surface within the FACE target area (79 samples). All samples were analyzed for silver using a sensitive method of atomic absorption spectrometry. The most important environmental result of the subprogram is that regardless of the manner of data stratification, even including the one extraneously high aircraft data point, the mean silver concentration of rainwater collected on seed days during the summer of 1973 is *at least* one order of magnitude below the current U. S. Public Health safety limit of 5×10^{-8} g ml^{-1}. Only one rainwater sample out of 69 collected by aircraft on seeded days contained a silver concentration exceeding this limit, and no other seeded sample was found to contain a silver concentration in excess of 3×10^{-9} g ml^{-1}. The median silver concentration from aircraft samples on seeded days was approximately 5×10^{-11} g ml^{-1}. The removal of the one extraneously high data point (1.4×10^{-7} g ml^{-1}) from the seeded day aircraft sample decreases the mean value of silver concentration by more than an order of magnitude to about 1×10^{-10} g ml^{-1}.

Of 58 rainwater samples collected by aircraft on nonseeded days, two contained a silver concentration in excess of 5×10^{-8} g ml^{-1} and three other silver concentration values exceeded 5×10^{-9} g ml^{-1}. The mean and median values of silver concentration from all 58 nonseeded samples were found to be approximately 8×10^{-9} and 1×10^{-10} g ml^{-1}, respectively. The admittedly arbitrary exclusion of three data points with silver values in excess of 1×10^{-8} g ml^{-1} results in a mean silver concentration for the 55 remaining samples of about 5×10^{-10} g ml^{-1}. The difference between the nonseeded sample mean of 5×10^{-10} g ml^{-1} and the seeded sample mean of 1×10^{-10} g ml^{-1} is significant at the 99% level using a Mann-Whitney Wilcoxon test. Note the rather surprising result that the mean silver concentration obtained by aircraft on nonseeded days is higher than that obtained on seeded days.

Of the 79 samples collected on the ground, no concentration of silver in excess of 1×10^{-9} g ml^{-1} was found, even though some mobile samples were obtained in rainshafts almost directly underneath treated clouds. The mean silver concentration from the surface samples collected on either seed or no-seed days was found to be three orders of magnitude lower than the U. S. Public Health's threshold of 5×10^{-8} g ml^{-1}. No significant differences were found between the seeded and nonseeded sets of surface data. This is particularly important in view of the fact that the NHEML experimental procedure requires a "massive" dynamic seeding approach with some seeded clouds receiving as much as 1 kg of silver iodide nucleant within a 10 min span.

The Coral Gables data set (Table 3) indicates a higher concentration of silver in precipitation at the surface during the course of the experiment then either before or after the experiment. It is somewhat surprising that this was so clearly evident at a location generally upwind of the target area. The mean concentration of silver in rainwater at the Coral Gables collection site during the experiment did not appear to differ greatly from that obtained at the surface within the target area.

Other conclusions to be drawn from the silver analysis are less clear-cut, with a high degree of variability (see Table 1) resulting in a somewhat confusing overall picture. The magnitudes of the silver concentration from surface rainwater data are comparable with those from surface collection programs carried out by various groups in other locations. The aircraft results, however, are puzzling in two respects: first, the silver concentrations appear consistently higher than those obtained on the ground and second, the nonseeded data set shows considerably more silver than does the seeded set. Several possible explanations for these results have been postulated in the text. They include 1) different collection methods, 2) different types of days, 3) collection of aerosol by the scoop, 4) possible contamination during sampling or preparation for analysis, and 5) persistence of silver iodide in the air mass for several days following seeding. It is clear, however, that additional data will be needed to resolve these uncertainties.

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